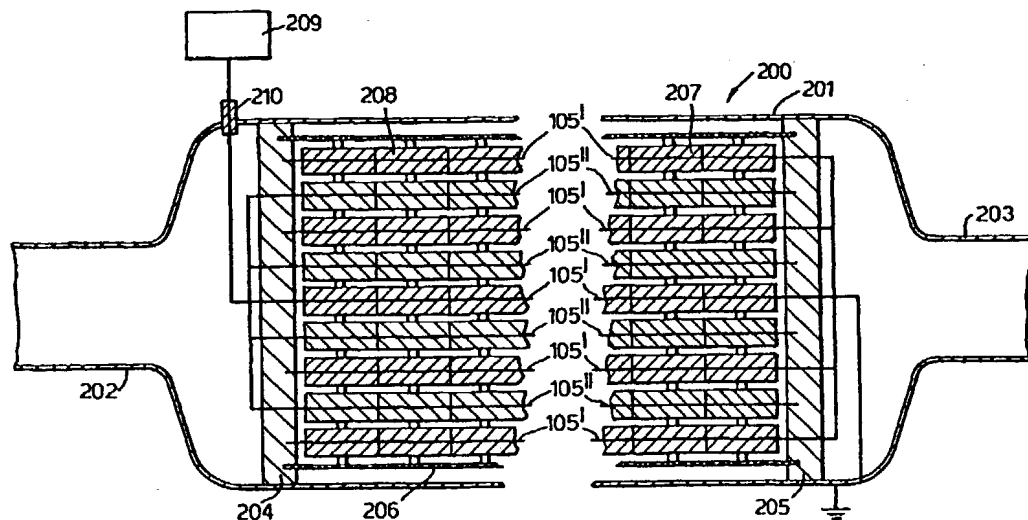




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(21) International Application Number: PCT/GB00/00397 (22) International Filing Date: 10 February 2000 (10.02.00) (30) Priority Data: 9903400.1 16 February 1999 (16.02.99) GB (71) Applicant (for all designated States except US): AEA TECHNOLOGY PLC [GB/GB]; 329 Harwell, Didcot, Oxfordshire OX11 0RA (GB). (72) Inventors; and (75) Inventors/Applicants (for US only): ANDREWS, Peter, James [GB/GB]; 17 Donnington Place, Belmont Park, Wantage, Oxfordshire OX12 9YE (GB). HALL, Stephen, Ivor [GB/GB]; 9 Blenheim Lane, Freeland, Oxford, Oxfordshire OX8 8AW (GB). INMAN, Michael [GB/GB]; 2 Longfellow Drive, Abingdon, Oxfordshire OX14 5NU (GB). SHAWCROSS, James, Timothy [GB/GB]; 27 Ticknell Piece Road, Charlbury, Oxfordshire OX7 3TN (GB). WEEKS, David, Michael [GB/GB]; 60 Blandy Avenue, Southmoor, Abingdon, Oxfordshire OX13 5DB (GB). MANSON-WHITTON, Christopher, David, John [GB/GB]; 19A Hillview Road, Botley Road, Oxford, Oxfordshire OX2 0DA (GB).		(74) Agents: LOFTING, Marcus, John et al.; AEA Technology plc, Patents Dept., 329 Harwell, Didcot, Oxfordshire OX11 0RA (GB). (81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published With international search report.

(54) Title: REACTOR FOR PLASMA ASSISTED GAS PROCESSING



(57) Abstract

A reactor for the plasma-assisted processing of gaseous media comprising a reactor chamber (200) including a gas permeable bed (100) of an active material, means (209) for applying across the bed of active material, a potential sufficient to establish a plasma in a gaseous medium flowing through the bed of active material and means (201, 202, 203) for constraining the gaseous medium to flow through the bed of active material, wherein the bed of active material comprises a matrix (207) of beads (103, 208) of a dielectric material consisting of an assembly of regular arrays of beads in each of which adjacent beads are connected to a high voltage input terminal or an earth point. (105', 105'').

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Reactor for Plasma Assisted Gas Processing

The present invention relates to reactors for the plasma-assisted processing of gaseous media and in particular to such reactors for the reduction of the emission of carbonaceous and nitrogenous oxide combustion products from the exhausts of internal combustion engines.

One of the major problems associated with the development and use of internal combustion engines is the noxious exhaust emissions from such engines. Two of the most undesirable materials, particularly in the case of diesel engines, are particulate matter (primarily carbon) and oxides of nitrogen (NO_x). Increasingly severe emission control regulations are forcing internal combustion engine and vehicle manufacturers to find more efficient ways of removing these materials in particular from internal combustion engine exhaust emissions. Unfortunately, in practice, it is found that combustion modification techniques which improve the situation in relation to one of the above components of internal combustion engine exhaust emissions tend to worsen the situation in relation to the other. A variety of systems for trapping particulate emissions from internal combustion engine exhausts have been investigated, particularly in relation to making such particulate emission traps capable of being regenerated when they have become saturated with particulate material.

30

Examples of such diesel exhaust particulate filters are to be found in European patent applications EP 0 010 384; US patents 4,505,107; 4,485,622; 4,427,418; and 4,276,066; EP 0 244 061; EP 0 112 634 and EP 0 132 166.

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In all the above cases, the particulate matter is removed from diesel exhaust gases by a simple, physical trapping of particulate matter in the interstices of a porous, usually ceramic, filter body, which is then
5 regenerated by heating the filter body to a temperature at which the trapped diesel exhaust particulates are burnt off. In most cases the filter body is monolithic, although EP 0 010 384 does mention the use of ceramic
10 beads, wire meshes or metal screens as well. US patent 4,427,418 discloses the use of ceramic coated wire or ceramic fibres.

In a broader context, the precipitation of charged particulate matter by electrostatic forces also is known.
15 However, in this case, precipitation usually takes place upon large planar electrodes or metal screens.

GB patent 2,274,412 discloses a method and apparatus for removing particulate and other pollutants from
20 internal combustion engine exhaust gases, in which the exhaust gases are passed through a bed of charged pellets of material, preferably ferroelectric, having high dielectric constant. In addition to removing
particulates by oxidation, especially electrical
25 discharge assisted oxidation, there is disclosed the reduction of NO_x gases to nitrogen, by the use of pellets adapted to catalyse the NO_x reduction.

A problem which arises with plasma assisted gas
30 processing reactors which include a bed of pellets of a high-dielectric constant material, such as those exemplified in specification GB 2 274 412, is that localised variations in the electric field in the pellet
bed can occur, possibly leading to regions of the pellet
35 bed in which the electric field is insufficient to enable

a plasma to be established in a gaseous medium flowing through the pellet bed of the reactor.

It is an object of the present invention to provide
5 an improved reactor for the plasma-assisted processing of gaseous media.

According to the invention there is provided a reactor for the plasma-assisted processing of gaseous
10 media, comprising a reactor chamber including a gas permeable bed of an active material, means for constraining a gaseous medium to flow through the bed of active material, wherein the bed of active material comprises a matrix array of first components of
15 dielectric material interspersed with second components of dielectric material, one or more first electrically conducting member or members electrically connected to a first electrical supply terminal being in electrical contact with at least one of the said first components,
20 one or more second electrically conducting member or members electrically connected to a second electrical supply terminal being in contact with all or a plurality of the said second components. In use a power supply is connected to apply an electrical potential across the
25 first and second electrical supply terminals, the potential and the arrangement of the array being such as to establish a plasma in a gaseous medium flowing through the bed of active material. In practice, it will be appreciated, one of the electrical supply terminals will
30 be connected to earth whilst the other is connected to a high voltage input supply.

Each of the first and second electrically conducting members may comprise a connecting wire or plate or a
35 combination of wire and plate. The dielectric constant or permittivity of the first and second components is

selected so as to optimise the plasma-assisted processing of the gaseous media flowing through the bed. An intimate contact between the electrically conducting members and their associated components of dielectric material is preferred to avoid plasma formation in any voids therebetween. The surface of the components of dielectric material in contact with the associated electrically conducting member may be coated with a metallic or other conducting coating to optimise this contact and prevent plasma formation therebetween and thus increase the electrical efficiency of the reactor for processing of the gaseous media.

In one embodiment of the invention the components of dielectric material are in the form of beads. Any one or a mixture of a variety of shapes may be adopted for the beads. Alternatively the components of dielectric material may be in the form of plates. The dielectric strength of the bead material is important in determining the size of bead that can be used in order to avoid electrical breakdown through the body of the dielectric. The higher the dielectric strength the smaller the bead size which can be used.

In another embodiment a dielectric coating is applied on electrically conducting members in the form of a wire or rod or plate, the coating thereby forming the component of dielectric material in situ. Such a coating may be deposited by a variety of methods including thermal spraying, for example by plasma-spraying as well as by wet chemical techniques for example by sol-gel processing. Dielectric material in the form of beads, coating or plates can have catalytic properties for processing of gaseous media. This may be particularly useful when for example processing nitrogeneous oxides and hydrocarbons in internal combustion engines as

described in our publication WO99/12638 and the specification of our application PCT/GB00/00079. Reductant gases such as hydrocarbon or nitrogen-containing reductant as described in PCT/GB00/00079 can be introduced before, after or into the reactor for processing of gaseous media. The reactor may be incorporated in a complete emission control system, in which emission control system catalyst material and reductant gases are utilised separately from the reactor.

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Preferably, the matrix array comprises first and second components of dielectric material arranged in alternate rows.

15 Examples of materials for the components of dielectric material include the aluminas known as LD 350, CT 530, Condea hollow extrudates, DYPAC, T-60 Alumina, T-162 alumina cordierite, α , χ and γ aluminas, and aluminas containing mixtures of these phases; ferroelectric materials such as titanates, particularly barium titanate; titania, particularly in the anatase phase; zirconia, vanadia, silver aluminate, perovskites, spinels, metal-doped zeolites and mixtures of these compounds. Examples of zeolites are those known as ZSM-5, Y, beta, mordenite all of which may contain iron, cobalt or copper with or without additional catalyst promoting cations such as cerium and lanthanum. Other examples of zeolites are alkali metal containing zeolites such as sodium Y zeolites. Examples of perovskites are La_2CuO_4 , $\text{La}_{1.9}\text{K}_{0.1}\text{Cu}_{0.95}\text{V}_{0.05}\text{O}_4$ and $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$. Vanadates including metavanadates and pyrovanadates such as potassium metavanadate, caesium metavanadate, potassium pyrovanadate and caesium pyrovanadate are also examples of dielectric materials. Selection of the permittivity is a parameter for optimisation of the plasma process. For example low permittivity material such as aluminium

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oxide or zeolite can be used for some plasma processing and higher permittivity material such as barium titanate for others.

5 The invention will now be described, by way of example, with reference to the accompanying drawings, in which

Figure 1 is a pictorial view of part of a plasma-
10 assisted gas reactor bed of active material embodying the invention,

Figure 2 is a longitudinal section of a schematic plasma-assisted gas reactor embodying the bed of active
15 material shown in Figure 1, and

Figure 3 is a longitudinal section of a second gas reactor embodying a bed of active material such as that shown in Figure 1.

20

Referring to Figure 1, a bed 100 of active material for a reactor for the plasma-assisted processing of gaseous media consists of a matrix 101 made of stacked identical arrays 102 of cylindrical beads 103 separated
25 by spacers 104. (Only two arrays 102 are shown). Corresponding beads of each array 102 are connected together by a plurality of parallel wires 105' and 105''.

The wires 105' are connected to a common high voltage input terminal (not shown in Figure 1) and the wires
30 marked 105'' are connected to a common earth point (also not shown in Figure 1). It should be appreciated that the spacers (104) separating the beads (103) are preferably of a lower permittivity than the beads themselves to promote the plasma-assisted processing of
35 gaseous media. This effect may also be achieved either

by making the spacers as thin as possible with respect to the bead or by extending the bead lengthwise along the electrode (105', 105'') while keeping the thickness of the spacer to a minimum. In another embodiment of the reactor shown in Figure 1 the spacers (104) can be removed from the arrays (102). In this example the wires (105') are connected to a common high voltage input location plate and the wires marked (105'') are connected to a common earth point location plate. The purpose of the plates is to provide an electrical connection to the electrodes within the assembly and to maintain the geometric shape and the parallel nature of the conductors.

Figure 2 shows such a reactor bed in a reactor for the plasma-assisted treatment of the exhaust gases of an internal combustion engine (not shown in the Figure) to reduce noxious emissions therefrom. Referring to Figure 2, the reactor 200 consists of a stainless steel reactor chamber 201 which has an inlet stub 202 and an outlet stub 203 by means of which it can be incorporated into the exhaust system of an internal combustion engine (not shown in the drawing). Inside the reactor chamber 201 are two support members 204 and 205 made of a ceramic insulating material such as alumina or MICATHERM micaceous glass as described in our publication WO99/20373. Housed into the supports 204 and 205 is a stainless steel cylinder 206 within which there is a matrix 207, such as that described with reference to Figure 1, of cylindrical beads 208 made of a dielectric material, the permittivity of which is selected so as to optimise the plasma-assisted processing of the gaseous media that is flowing through the reactor.

The connecting wires 105'' of one set of beads 208

pass through the support 204 and are connected to a source of pulsed high potential 209 via a high voltage lead-through 210. The connecting wires 105' of the other set of beads 208 pass through the support 205 and thence
5 to an earth point 211 to which the reactor casing 201 and the stainless steel cylinder 206 also are connected. The regions of the supports 204, 205 within the inner cylinder 206 are made to be readily gas permeable so as to keep to a minimum the back-pressure introduced into
10 the exhaust system by the reactor. In another embodiment the electrodes in the form of wires (105') and (105'') may be replaced by electrodes in the form of flat conducting plates and the beads (208) are then replaced by sheets of dielectric material. Such a configuration can be
15 fabricated by applying to the flat electrode plates a coating of a dielectric material by thermal spraying, for example by plasma-spraying or by wet chemical techniques for example by sol-gel processing. The matrix (207) will then be a series of stacked arrays consisting of flat
20 conducting high voltage and earth plates sandwiched between dielectric material in the form of sheets in intimate contact with the plate. Alternatively a metallic electrode can be deposited onto a dielectric plate or wafer or rod by any of a variety of techniques including
25 chemical vapour deposition, thermal spraying, wet chemical techniques, screen printing, painting, dipping or other similar technique. Metal-coated dielectric materials are assembled in such a way that the metal is enclosed and in contact with dielectric material. In
30 another embodiment some of the stacked arrays in the reactor can be replaced by catalyst material in the form of spheres, pellets, extrudates, fibres, sheets, coils, granules, foam or honeycomb monolith or as a coating on a ceramic foam or ceramic honeycomb.

Figure 3 shows a reactor for the plasma-assisted treatment of the exhaust gases such as those from an internal combustion engine to reduce noxious emissions therefrom, which is configured to provide a substantially radial flow of exhaust gases through the reactor bed, instead of the axial flow regime of the embodiment of Figure 1. Those components which are common to both embodiments have the same reference numerals.

Referring to Figure 3, the majority of the two reactors are the same. The insulating supports 204, 205, however, are replaced by similar supports 301, 302 which are not gas permeable and the matrix 207 of beads 208 is contained between two co-axial cylinders 303, 304 made of perforated stainless steel, both of which are housed into the supports 301, 302. The support 301 has a series of regularly spaced axial holes 305 around its periphery which open into the space 306 between the stainless steel cylinder 303 and the reactor casing 201. The support 302 on the other hand has a hole 307 in its centre. The diameter of the hole 307 in the support 302 is approximately the same as the internal diameter of the inner stainless steel cylinder 304. Exhaust gases entering the reactor chamber 201 are, therefore, constrained to flow radially through the bed 207 of beads 208 before leaving the reactor chamber 201 via the hole 307 in the centre of the support 302.

The power supply 209 is adapted to produce pulses having a potential of the order of kilovolts to tens of kilovolts and repetition frequencies in the range 50 to 5000 Hz, although higher frequencies of the order of tens of kilohertz can be used. Pulsed direct current is convenient for automotive use, but alternating potentials for example triangular or sine waves of the same or similar characteristics can be used. The potential

applied is selected according to the design of the plasma reactor and to optimise the plasma-assisted processing of the gaseous media..

- 5 It will be appreciated that the power supply may advantageously be located adjacent to the reactor as described in our publication WO99/05400 and the specification of our application PCT/GB00/00108.

Claims

1. A reactor for the plasma-assisted processing of
5 gaseous media, comprising a reactor chamber (200)
including a gas permeable bed (100) of an active
material, means (201,202,203) for constraining a gaseous
medium to flow through the bed of active material,
wherein the bed of active material comprises a matrix
10 array (207) of components (103;208) of dielectric
material, characterised in that the matrix array
comprises first components (103;208) of dielectric
material interspersed with second components (103;208) of
dielectric material, one or more first electrically
15 conducting member or members (105') electrically
connected to a first electrical supply terminal being in
electrical contact with at least one of the said first
components, one or more second electrically conducting
member or members (105'') electrically connected to a
20 second electrical supply terminal being in contact with
all or a plurality of the said second components.
2. A reactor as claimed in claim 1, further
characterised in that a power supply is connected to
25 apply an electrical potential across the first and second
electrical supply terminals, the potential and the
arrangement of the array being such as to establish a
plasma in a gaseous medium flowing through the bed (100)
of active material.
- 30
3. A reactor as claimed in claim 1 or claim 2, further
characterised in that each of the first and second
electrically conducting members (105';105'') comprises a
connecting wire or plate or a combination of wire and
35 plate.

4. A reactor as claimed in any one of the preceding claims, further characterised in that the electrically conducting members (105';105'') and their associated components (103;208) of dielectric material are in
5 intimate contact to avoid plasma formation in any voids therebetween.

5. A reactor as claimed in claim 4, further characterised in that surfaces of the components
10 (103;208) of dielectric material in contact with the associated electrically conducting member (105';105'') are coated with a metallic or other conducting coating to optimise this contact and prevent plasma formation therebetween.

15 6. A reactor as claimed in claim 4, further characterised in that the electrically conducting members (105';105'') are formed by deposition onto their associated component (103;208) of dielectric material.

20 7. A reactor as claimed in any preceding claim, further characterised in that the electrically conducting members (105';105'') are enclosed in and surrounded by dielectric material.

25 8. A reactor as claimed in any preceding claim, further characterised in that the components (103;208) of dielectric material are in the form of beads.

30 9. A reactor as claimed in claim 8, further characterised in that the beads (103) are separated by spacers (104).

35 10. A reactor as claimed in claim 8 or claim 9, further characterised in that all the beads (103) are the same size.

11. A reactor as claimed in claim 8 or claim 9 or claim 10, further characterised in that the beads (103) are cylindrical or spherical in shape.

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12. A reactor as claimed in any one of claims 1 to 7, further characterised in that the components (103;208) of dielectric material are in the form of plates.

10 13. A reactor as claimed in any one of claims 1 to 5, further characterised in that a dielectric coating is applied on the electrically conducting members (105';105''), which are in the form of a wire or rod or plate, the coating thereby forming the component
15 (103;208) of dielectric material in situ.

14. A reactor as claimed in any of the preceding claims, further characterised in that the matrix array comprises first and second components (103;208) of dielectric
20 material arranged in alternate rows.

15. A reactor as claimed in any of the preceding claims, further characterised in that the matrix array is monolithic.

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16. A reactor as claimed in any of the preceding claims, adapted to be connected into the exhaust system of an internal combustion engine.

30 17. A reactor as claimed in any of the preceding claims in association with a power supply adapted to produce pulsed dc potentials of the order of kilovolts or tens of kilovolts and with repetition frequencies of the order of kilohertz.

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18. A reactor as claimed in any of the preceding claims

incorporated in a complete emission control system for an internal combustion engine, in which catalyst material and reductant gases are utilised separately from the reactor.

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19. A reactor as claimed in any of the preceding claims, in or in association with which catalyst material is utilised, the catalyst material being selected from one or a mixture of the following: aluminas, titanias
10 zirconia, vanadia, silver aluminate, perovskites such as La_2CuO_4 , $\text{La}_{1.9}\text{K}_{0.1}\text{Cu}_{0.95}\text{V}_{0.05}\text{O}_4$ and $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$, spinels, metal-doped zeolites such as ZSM-5, Y, beta, mordenite all of which zeolites may contain iron, cobalt or copper with or without additional catalyst promoting cations
15 such as cerium and lanthanum, alkali metal containing zeolites such as sodium Y zeolites, vanadates such as metavanadates and pyrovanadates including potassium metavanadate, caesium metavanadate, potassium pyrovanadate and caesium pyrovanadate.

20

20. A reactor as claimed in claim 19, further characterised in that the catalyst material is in the form of spheres, pellets, extrudates, fibres, sheets, coils, granules, foam or honeycomb monolith or as a
25 coating on a ceramic foam or ceramic honeycomb.

21. A reactor as claimed in any of the preceding claims, further characterised in that the said matrix array includes catalyst material in the array interspersed
30 between said first components and/or said second components.

Fig.1.

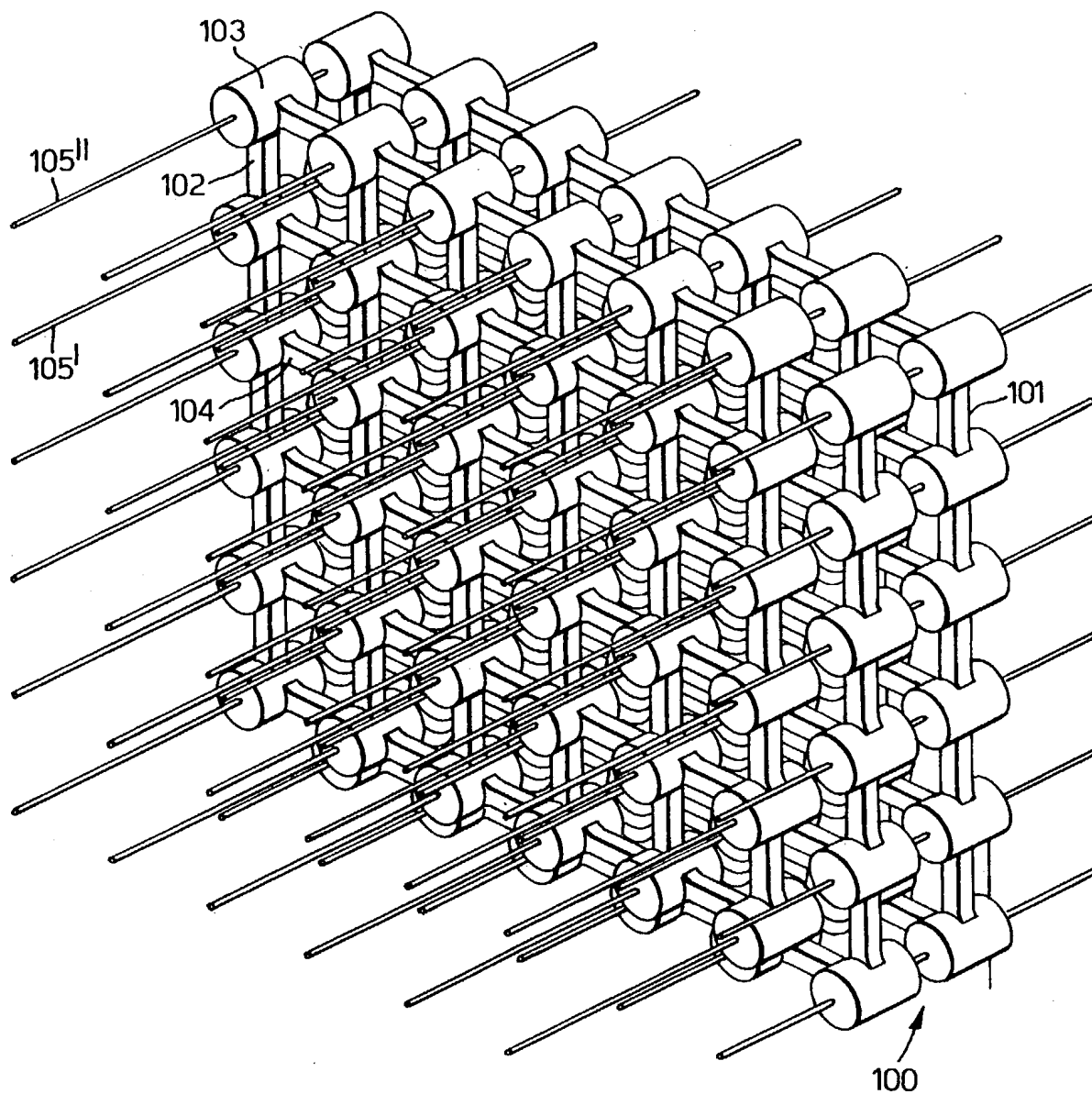
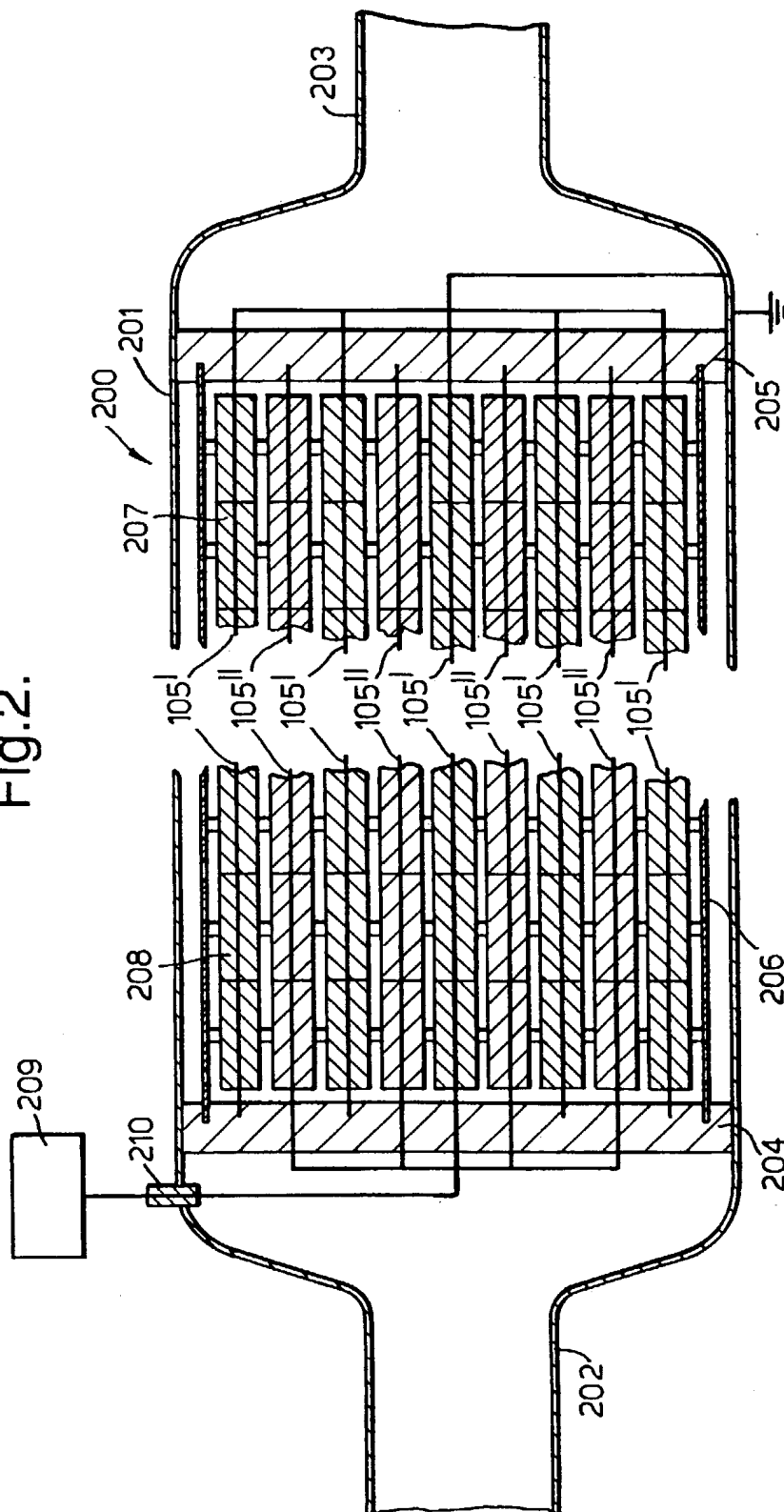
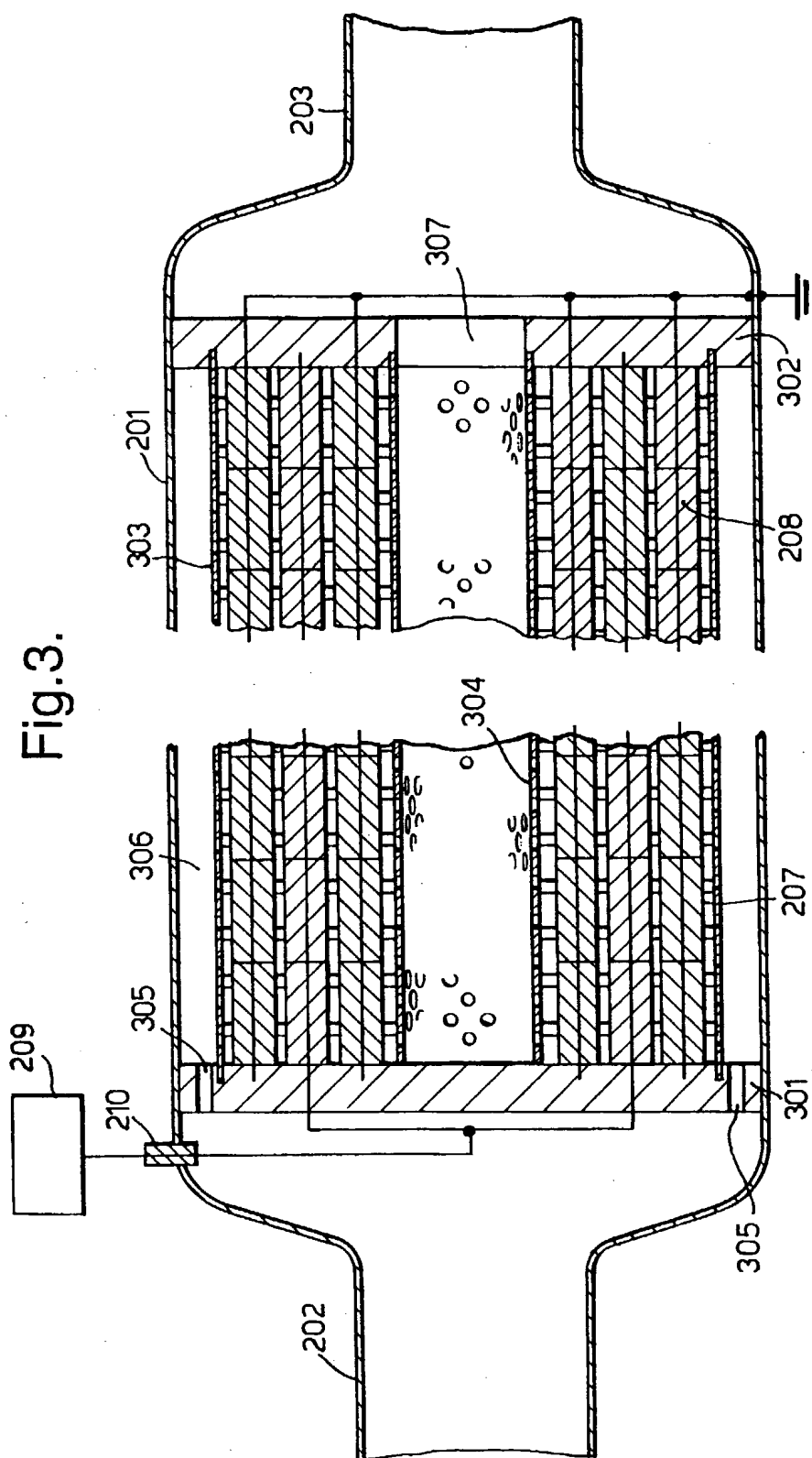


Fig.2.



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INTERNATIONAL SEARCH REPORT

Int. Application No

PCT/GB 00/00397

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 F01N3/08 B01D53/32 F01N3/01

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 F01N B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 99 47242 A (FRENTON LTD ;BELENKOV EDUARD LEONIDOVICH (RU); GUDKOV IGOR ALEXAND) 23 September 1999 (1999-09-23) page 15, line 28 -page 17, line 4 page 19, line 29 -page 20, line 9 figures 2,3	1-5,7, 12-14, 16,17
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	-/-	



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Patent family members are listed in annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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